

# Negative density of states: screening, Einstein relation, and negative diffusion.

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## Abstract

In strongly interacting electron systems with low density and at low temperature the thermodynamic density of states is negative. It creates difficulties with understanding of the Einstein relation between conductivity and diffusion coefficient. Using the expression for electrochemical potential that takes into account the long range part of the Coulomb interaction it is shown that at negative density of states Einstein relation gives a negative sign of the diffusion coefficient  $D$ , but under this condition there is no thermodynamic limitation on the sign of  $D$ . It happens because the unipolar relaxation of inhomogeneous electron density is not described by the diffusion equation. The relaxation goes much faster due to electric forces caused by electron density and by neutralizing background. Diffusion coefficient is irrelevant in this case and it is not necessarily positive because process of diffusion does not contribute to the positive production of entropy. In the case of bipolar diffusion negative  $D$  results in a global absolute instability that leads to formation of neutral excitons. Graphene is considered as an example of a system, where the density relaxation is expected to be due to electric force rather than diffusion. It may also have a negative density of states.

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## I. INTRODUCTION

The idea of the Einstein relation was put forward by Einstein<sup>1</sup> and Smoluchowski<sup>2</sup> in 1905-1906. Both scientists considered the Brownian motion in the presence of gravitational force. The result is the relation between mobility  $u$  in the field and diffusion coefficient  $D$ . In case of electric field and particles with the charge  $e$  it has a form

$$eD = Tu, \quad (1)$$

where  $T$  is the temperature in energy units. The main idea was equivalence of an external force and the density gradient. Of course, both Einstein and Smoluchowski did not care about negligible mutual gravitational or any other small interactions of the Brownian particles.

The formulation of the Einstein relation for electrons is based upon electrochemical potential, the thermodynamic function that, like temperature and pressure, should be the same at all points of the system in the equilibrium state. The usual arguments are as follows. If an external potential  $\psi$  is applied to the system, the condition of thermodynamic equilibrium reads

$$E_{ec} = \mu(n) + e\psi = \text{Const}, \quad (2)$$

where  $\mu(n)$  is the chemical potential as a function of inhomogeneous electron density  $n$ . In the equilibrium both  $n$  and  $\psi$  are function of coordinates while  $E_{ec}$  is constant. The temperature  $T$  should also be constant. Therefore, the electrical current density  $\mathbf{j}$  at constant  $T$  can be written in a form<sup>3</sup>

$$\mathbf{j} = -\frac{\sigma}{e} \nabla E_{ec} = \sigma \mathbf{E} - D \nabla en, \quad (3)$$

where  $\sigma$  is conductivity and  $\mathbf{E} = -\nabla\psi$ . Then one gets relation connecting  $\sigma$  and  $D$

$$\frac{\sigma}{e^2} \frac{d\mu}{dn} = D, \quad (4)$$

which is also called Einstein relation. For the Boltzman gas  $d\mu/dn = T/n$  and one gets Eq.(1) if  $\sigma = enu$ . It looks like derivation of Eq. (4) is independent of the properties of the system and this equation can be consider as general thermodynamic law.

A simple observation shows however that in the case of non-ideal electron gas the Einstein relation needs some comments. We discuss an electron gas on the positive background at

low temperatures and low densities when dimensionless parameter  $r_s$  is not very small. Here  $r_s^3 = 3/(4\pi n a_B^3)$  for 3-d case and  $r_s^2 = 1/\pi n_2 a_B^2$ , where  $n$  and  $n_2$  are 3- and 2-dimensional electron densities respectively and  $a_B = \hbar^2 \kappa / m e^2$  is the Bohr radius,  $m$  is an effective electronic mass,  $\kappa$  is an effective permittivity.

The problems of dynamic screening and diffusion in slightly non-ideal electron gas ( $r_s \ll 1$ ) with electron-electron interaction were considered in details about 20 years ago (See Ref.[<sup>4,5,6</sup>]) In this case the thermodynamic density of states is large and positive. I concentrate here on the strongly non-ideal case  $r_s \geq 1$ .

An electron gas on the positive background at low temperatures and low densities has energy  $E$  of the order of  $-e^2 n^{1/d} N / \kappa$ , where  $d = 2, 3$  is the space dimensionality and  $n$  is the density per area or volume respectively,  $N$  is total number of electrons. Then  $\mu \sim -e^2 n^{1/d} / \kappa$  and  $E, \mu$ , and  $d\mu/dn$  are negative<sup>7,8</sup>. The first experimental confirmation of this idea was done by Kravchenko *et al*<sup>9,10</sup>, but direct quantitative study of this effect was performed by Eisenstein *et al*<sup>11,12</sup>.

The derivative  $d\mu/dn$  is proportional to the reciprocal compressibility of the electron gas. Note that compressibility has to be positive due to the thermodynamical condition of stability. However, this principle cannot be applied to the charged systems, like electron gas, because part of their energy is outside the system in a form of the energy of electric field. On the other hand, in the case of a neutral electron-hole plasma, the situation of negative compressibility can arise leading to collapse of the system. Such a situation is considered at the end of Sec. III.

It follows from Eq. 4 that if  $d\mu/dn$  is negative, diffusion coefficient  $D$  and conductivity  $\sigma$  have opposite signs. This observation needs an explanation because near the thermodynamic equilibrium both of them have to be positive to provide positive entropy production due to the Joule heat and due to the relaxation of inhomogeneous density.

## II. ELECTROCHEMICAL POTENTIAL AND STATIC SCREENING

To resolve this contradiction one should include the long-range part of the Coulomb potential created by inhomogeneous electron gas into the function  $E_{ec}$  in Eq. (2). This contribution is a functional of  $n(\mathbf{r})$ .

To find  $E_{ec}$  taking into account electron-electron interaction one should minimize the

Helmholtz energy  $F$  with respect to electron density  $n(\mathbf{r})$  at a given value of  $T$  and  $N$ . For low  $T$  one gets

$$F = \frac{e^2}{2\kappa} \int \int \frac{n'(\mathbf{r})n'(\mathbf{r}')d^3rd^3r'}{|\mathbf{r} - \mathbf{r}'|} + \int f(n + n')d^3r + \int en'(\mathbf{r})\psi d^3r - E_{ec} \int n'(\mathbf{r})d^3r, \quad (5)$$

where  $f$  is the Helmholtz energy density of a homogeneous electron system that results from the interaction in a neutral system, like the Wigner crystal or "Wigner liquid". Since this interaction comes mainly from the nearest neighbors and  $n(\mathbf{r})$  is a smooth function, one may assume that both  $f$  and chemical potential  $\mu = df/dn$  are local functions of  $n(\mathbf{r})$ . We assume also that  $n(\mathbf{r}) = n + n'(\mathbf{r})$ , where  $n$  is average density and  $n' \ll n$ .

Minimization of this expression with respect to  $n'$  gives the equation

$$E_{ec} = \mu(n) + e\psi + \frac{d\mu}{dn}n' + \frac{e^2}{\kappa} \int \frac{n'(\mathbf{r}')d^3r'}{|\mathbf{r} - \mathbf{r}'|}. \quad (6)$$

It differs from Eq. (2) by the potential of electrons in the right hand side. Note that this potential is due to the violation of neutrality in a scale much larger than the average distance between electrons. To check this equation we consider thermodynamic equilibrium and find equations for the Thomas-Fermi static screening in 3- and 2-dimensional cases. Since  $E_{ec}$  is independent of  $\mathbf{r}$  in thermodynamic equilibrium one may take  $E_{ec} - \mu(n)$  as a reference point for the total potential  $\varphi$  defined as

$$\varphi = \psi + \frac{e}{\kappa} \int \frac{n'(\mathbf{r}')d^3r'}{|\mathbf{r} - \mathbf{r}'|}. \quad (7)$$

It follows from Eq. (6) that

$$e\varphi = -\frac{d\mu}{dn}n'. \quad (8)$$

The Poisson equation has a form

$$\nabla^2\varphi = -\frac{4\pi(en' - \rho_{ext})}{\kappa}, \quad (9)$$

where  $\rho_{ext}$  is density of external charge. Using Eq. (8) one gets final equation for the 3-d linear screening

$$\nabla^2\varphi = -q_3^2\varphi - \frac{4\pi\rho_{ext}}{\kappa}. \quad (10)$$

Here

$$q_3^2 = \frac{4\pi e^2}{\kappa} \frac{dn}{d\mu} \quad (11)$$

is the reciprocal 3-dimensional screening radius.

Consider now a thin layer (x-y plane) with 2d electron gas separating two media with dielectric constants  $\kappa_1$  and  $\kappa_2$ . In this case one should substitute  $n \Rightarrow n_2\delta(z)$  and  $\kappa \Rightarrow \bar{\kappa} = (\kappa_1 + \kappa_2)/2$ . The results is<sup>13</sup>

$$\nabla^2\varphi = -q_2\varphi\delta(z) - \frac{4\pi\rho_{ext}}{\bar{\kappa}}, \quad (12)$$

where

$$q_2 = \frac{2\pi e^2}{\bar{\kappa}} \frac{dn_2}{d\mu}. \quad (13)$$

It is important that Eqs. (10), (12) are applicable only if the screening is linear ( $n' \ll n$ )<sup>14</sup>. There is another serious problem of applicability the Thomas-Fermi approximation in the case of the negative density of states. Indeed, the dielectric permittivity in this approximation has a form

$$\epsilon(q) = \kappa(1 - \frac{|q_3|^2}{q^2}) \quad (14)$$

in 3-d case and

$$\epsilon(q) = \kappa(1 - \frac{|q_2|}{q}) \quad (15)$$

in 2-d case. In both cases it has roots at  $q = |q_3|, |q_2|$ . The expression for the screened potential  $\varphi$  has a form

$$\varphi(\mathbf{r}) = \int \frac{\varphi_0(\mathbf{q}) \exp(i\mathbf{q} \cdot \mathbf{r}) d\mathbf{q}}{\epsilon(\mathbf{q})}, \quad (16)$$

where  $\varphi_0$  is a bare potential. Thus, the roots of  $\epsilon$  transform into the first order poles without any reasonable way of the detour. Such a detour follows from the causality for the  $\omega$ -plane but not for the  $q$ -plane. Moreover, the electrostatic potential should be real and one cannot add a small imaginary part in the denominator. Therefore I think that the poles do not have any physical sense.

The reason is that negative sign of the density of states appears when  $q_3, q_2$  are of the order of average distance between electrons  $\bar{r}$ . At such distances the very concept of macroscopic field does not have sense. However, if the bare potential has only harmonics with  $q \ll |q_3|, |q_2|$ , the Eqs.(10,12) have a sense. Consider, for example, the screening of the positive charge  $Z$  at a distance  $z_0$  from the plane with 2-d gas(plane  $z = 0$ ). The solution of Eq.(12) has a form<sup>13</sup>

$$\varphi(\rho) = \int_0^\infty \frac{Z \exp(-qz_0)}{\kappa(q + q_2)} q J_0(q\rho) dq, \quad (17)$$

where  $\rho$  is a polar radius in the plane  $z = 0$ . Suppose that  $|q_2|z_0 \gg 1$ . Now the contribution to integral Eq.(17) from  $q \simeq |q_2|$  is exponentially small and one can ignore  $q$  in the denominator. Then

$$\varphi(\rho) = \frac{Zz_0}{\kappa q_2 (z_0^2 + \rho^2)^{3/2}}. \quad (18)$$

Note that at  $q_2 < 0$  a positive charge creates a small *negative* potential in the plane with electrons. That is what I call "overscreening".

Extra electron density, as calculated from Eq. (8) is

$$en' = -\frac{Zz_0}{2\pi(z_0^2 + \rho^2)^{3/2}} \quad (19)$$

It is negative and independent of the sign of  $q_2$ . One can see that the total charge

$$\int_0^\infty en' 2\pi\rho d\rho = -Z \quad (20)$$

Due to geometry of the problem electric field is zero below the plane with electrons. As follows from Eq. (8), the signs of charge density and potential are opposite if the density of states is negative.

For the case of two such planes (double quantum well structure) Luryi<sup>15</sup> has predicted a small penetration of electric field through the first plane. He has considered the case of positive density of states. Then the small penetrating field between two planes has the same direction as the incident field.

Eisenstein *at al.*<sup>11</sup> studied this effect experimentally and found out that at negative density of states the propagating field is opposite to the incident field and this is also a result of the overscreening (see the quantitative theory in Ref.<sup>12,16,17</sup>).

Negative density of states was also used<sup>18</sup> for the explanation of magnetocapacitance data by Smith *at al.*<sup>19</sup>.

### III. CONDUCTIVITY VERSUS DIFFUSION

Now I come back to the problem of the negative diffusion. If the system is not in equilibrium the electric current can be written in the same form as Eq. (3)

$$\mathbf{j} = -\frac{\sigma}{e}\nabla E_{ec}. \quad (21)$$

Using Eq. (6) one gets

$$\mathbf{j} = \sigma \mathbf{E} - D \nabla en' - \sigma \frac{e}{\kappa} \nabla \int \frac{n'(\mathbf{r}') d^3 r'}{|\mathbf{r} - \mathbf{r}'|}. \quad (22)$$

Here  $D$  is connected to  $\sigma$  by the Einstein relation Eq. (4). Considering relaxation of the charge density one can ignore external field  $\mathbf{E}$ . The relaxation is described by the continuity equation

$$\frac{\partial(en)}{\partial t} = -\nabla \cdot \mathbf{j} \quad (23)$$

or

$$\frac{\partial(en)}{\partial t} = \sigma \left( \frac{1}{e^2} \frac{d\mu}{dn} \nabla^2(en') - \frac{4\pi en'}{\kappa} \right). \quad (24)$$

The ratio  $R$  of the first (diffusion) term in the right hand side to the second (field) term is  $R = (q_3^2 L^2)^{-1}$ , where  $L^{-2} = \nabla^2 n' / n'$  is the characteristic size of the extra charge and  $q_3^2$  is given by Eq. (11). If electron gas is non-ideal,  $q_3 \sim 1/\bar{r}$ , where  $\bar{r}$  is the average distance between electrons. However, the very concept of diffusion equation is valid at  $L \gg \bar{r}$ . This means that for the non-ideal gas  $|R| \ll 1$  and the diffusion term in Eq. (24) should be ignored. Then the equation has a simple solution

$$n'(\mathbf{r}, t) = n'(\mathbf{r}, 0) \exp(-(t/\tau_M)), \quad (25)$$

where  $\tau_M = \kappa/(4\pi\sigma)$  is well-known Maxwell's time. Coefficient  $D$  does not enter in this case in the entropy production and it does not have a physical sense. Thus in 3-dimensional non-ideal electron gas negative  $d\mu/dn$  does not create any contradiction with the Einstein relation.

In the 3d gas of high density  $\mu \sim n^{2/3}$  and  $R \sim (\bar{r}/L)^2/r_s$  with  $r_s < 1$ . In this case  $R$  might be large and diffusion is possible. However  $d\mu/dn > 0$ , and  $D > 0$ .

Now we consider the relaxation of the charge density in 2-dimensional case. Instead of Eq.(24) one gets

$$\begin{aligned} \frac{\partial(en_2)}{\partial t} = & \sigma_2 \left( \frac{1}{e^2} \frac{d\mu}{dn_2} \nabla^2(en'_2) \right. \\ & \left. - \frac{e}{\kappa} \nabla^2 \int \frac{n'_2(\mathbf{r}') d^2 r'}{|\mathbf{r} - \mathbf{r}'|} \right). \end{aligned} \quad (26)$$

Here  $n_2, \sigma_2$  and  $\nabla$  are 2-dimensional density, conductivity, and 2-dimensional gradient respectively. To consider the ratio  $R_2$  of the first (diffusion) term to the second (field) term it

is convenient to make the Fourier transformation. Then one gets

$$\frac{\partial(n_q)}{\partial t} = -\sigma_2 \left( \frac{1}{e^2} \frac{d\mu}{dn_2} q^2 n_q + \frac{2\pi q}{\bar{\kappa}} n_q \right), \quad (27)$$

where  $n_q$  is the Fourier transformation of  $n'_2$ .

Now we find that the ratio of the first (diffusion) term in the right hand side of Eq. (27) to the second (field) term  $R_2 = q/q_2$ , where  $q_2$  is given by Eq. (13). Similar to the 3d case in the non-ideal gas  $|q_2| \sim 1/\bar{r}$  and diffusion should be ignored. Then we get the Dyakonov-Furman equation<sup>20</sup>

$$\frac{\partial(n_q)}{\partial t} = -v q n_q, \quad (28)$$

where velocity  $v = 2\pi\sigma_2/\bar{\kappa}$ . The physical meaning of this equation is that extra density of electrons localized initially at some spot propagates in all directions with velocity  $v$  conserving the total amount of extra electrons. Of course, this way of relaxation is more efficient than diffusion (random walk), because  $r \sim vt$  while  $r \sim \sqrt{Dt}$  in the case of diffusion. Thus, diffusion coefficient  $D$  is irrelevant and negative  $d\mu/dn$  does not create any contradiction with the Einstein relation. In a high density electron gas  $R_2 = q\bar{r}/r_s$  and diffusion mechanism is possible. In this case  $d\mu/dn > 0$  and  $D > 0$ .

One can consider this problem from a different point of view. In both 3d and 2d cases the negative diffusion coefficient  $D$  appears in the term with the highest derivative that leads to the absolute instability even if  $D$  is small<sup>21</sup>. Consider, for example Eq. (24) for 3d case. After the Fourier transformation the solution for the charge density  $\rho = en'$  can be written in a form

$$\rho_q = \rho_q^0 \exp \left( -\frac{4\pi\sigma t}{\kappa} - Dq^2 t \right), \quad (29)$$

where  $D$  is given by the Einstein relation Eq. (4). One can see that at  $D < 0$  solution increases with time exponentially for harmonics with  $q\bar{r} \geq 1$ .

The physical explanation is as follows. The Eqs.(24,26) contain average distance between electrons  $\bar{r}$ . So they contain information that the charged liquid has a discrete electronic structure. This information comes from the negative density of states which originates from the interaction of the separate electrons. That is why macroscopic equations become unstable at small spacial harmonics. The message is that  $n(\mathbf{r})$  is rather a set of  $\delta$ -functions than a continuous function. The instability is absent if  $D$  is positive.

The instability of small spatial harmonics at small negative  $D$  does not affect larger harmonics because Eqs.(24,26) are linear. Due to the linearity different harmonics are in-



dependent and transformation of energy from small spacial harmonics to large harmonics is forbidden (cp. phenomenon of turbulence in non-linear hydrodynamics where the transformation of energy is not forbidden, but the instability is initiated by large harmonics).

Therefore, I think that at small  $D$  approximation  $D = 0$  that gives Eqs.(25,28) is correct.

One should note that the problem of the non-physical roots of electric permittivity discussed in the previous section is of the same nature.

Before we discussed the unipolar diffusion. Consider the simplest case of the ambipolar diffusion assuming that at  $t = 0$  the densities of electrons and holes are equal in some finite region of space and are zero otherwise. Moreover we assume that the local macroscopic charge density  $\rho(\mathbf{r}, t) = 0$  and a recombination of carriers is very slow. In this case Eq. (6) describes the electron-hole system in quasi-equilibrium. At large  $r_s$  one gets  $E, \mu, d\mu/dn < 0$  but the last term in Eq. (6) is absent. So the smearing of the density of particles is described by the equation of diffusion at all  $r_s$ , but at small density ( $r_s \geq 1$ ) coefficient  $D < 0$ . Then the absolute instability takes place for all harmonics that means a collapse of the system. Thus the electron-hole "Wigner liquid" and crystal are unstable.

This result is very transparent. It happens because negative  $\mu$  just means that the energy of the system decreases with increasing density. In bipolar case neutrality is provided by the particles and we do not consider any background. Thus the instability is a result of the negative compressibility in a neutral system. At large enough  $r_s$  these particles are classical, and the absence of the mechanical equilibrium follows also from the Earnshaw theorem. In reality quantum mechanics becomes more important with increasing density. As a result the excitons are formed. These neutral particles have a positive diffusion coefficient  $D_a$  and their density smears with time through all available space. This process is described by a regular diffusion equation. In the case of optical excitation the carriers may appear in the form of the excitons from the very beginning

For the coefficient of the ambipolar diffusion  $D_a$  a textbook equation<sup>22</sup>

$$D_a = \frac{2D_e D_h}{D_e + D_h} \quad (30)$$

is often used, where  $D_{e,h}$  are diffusion coefficients of electron and holes in unipolar case. As follows from the previous discussion, one should be careful with this equation because for the non-ideal electron (or hole) gas these unipolar coefficients might be negative and meaningless. It happens because in unipolar case there is a deviation from neutrality that

creates electric field, while in bipolar case the system is neutral. In this case Eq. (30) does not work and one should calculate  $D_a$  in a different way as a diffusion of the exciton.

In the recent paper by Zhao<sup>23</sup> the experimental results for the ambipolar diffusion in silicon-on-insulator system are compared with Eq. (30). At high temperatures a good agreement is found while at low temperatures the observed values of  $D_a$  are 6-7 times less. The previously reported values<sup>24</sup> show similar temperature dependence.

The author's explanation is that coefficients  $D_{e,h}$  are taken for the bulk silicon using Einstein relation and they might be larger than in the film at low temperatures. However, the reason discussed above cannot be excluded.

#### IV. GRAPHENE AS A POSSIBLE EXAMPLE OF A NON-IDEAL ELECTRON SYSTEM

It is interesting to discuss the single layer graphene as an example of the system with non-ideal electron gas. Graphene is a gapless material with the linear spectrum of electrons and holes near the Dirac point. Due to some reasons, that are not quite clear now, the velocity  $v$  of electrons and holes in equation  $\epsilon = \pm pv$  is of the order of  $e^2/\hbar$ . It follows that at any Fermi energy inside this linear spectrum electron gas in graphene is non-ideal in a sense mentioned above: the absolute value of the chemical potential is of the order of interaction energy  $e^2 n^{1/2}$ . It means that unipolar density relaxation in this system should be described by the Dyakonov-Furman equation rather than by diffusion equation.

However, without magnetic field the electron gas in graphene is marginally non-ideal. It cannot be classical, like an electron gas of a low density with quadratic spectrum. The marginal situation makes theoretical calculations very difficult. Nevertheless, it is accepted that the Wigner crystal in single layer graphene is absent without magnetic field<sup>25,26</sup>. The sign of  $d\mu/dn$  is also an interesting question but very difficult for theoretical study. Recently tunneling microscopy experiment has been done by Martin *et al.*<sup>27</sup>. They claim that their measurement give the thermodynamic density of states and that it is positive. The last statement might be a result of disorder.

## V. CONCLUSION

Finally I argue that the negative sign of diffusion coefficient that follows from the Einstein relation at negative density of states does not lead to any contradiction because diffusion coefficient is irrelevant for the unipolar transport under this condition. The sign of the diffusion coefficient in this case should not be definitely positive because the diffusion is not the main source of the entropy production. In bipolar situation negative diffusion means the collapse of the system and formation of neutral excitons.

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